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Solution Processable White Organic Light-Emitting Diodes Using New Blue Host Material Including Substituent Group

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New host material of T-TATa isomer substituted t-butyl group was investigated in solution process WOLED device compared with 4-(10-(3',5'-diphenylbiphenyl-4-yl)anthracen-9-yl)-*N*,*N*-diphenylaniline [TATa]. A two-color WOLED of a co-host system using solution process method was demonstrated. The device configuration was ITO/PEDOT:PSS (40 nm)/emitting layer (50 nm)/TPBi (20 nm)/LiF (1 nm)/Al. The emitting layer consisted of TATa or T-TATa isomer, NPB, DPAVBi (blue dopant), and rubrene (yellow dopant). NPB was used as not only blue host but also helping hole carrier transport. The device using T-TATa compound as a co-host exhibited a luminance efficiency of 3.39 cd/A, which is about twice higher than TATa device of 1.58 cd/A at 10 mA/cm².

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1. INTRODUCTION

Many researchers have extensively studied to make high performance organic light-emitting diode (OLED).¹⁻⁴ Especially, The interest based on white OLED (WOLED) has been rapidly growing in large full color display such as OLED TV and lighting fields.⁵ Until now, WOLED has been produced using vacuum evaporation method, but this method involves disadvantages such as high cost, difficulties of large scaling and adjusting doping concentration.^{6,7}

Therefore, there are many researches on methods of producing solution processed WOLED such as spin-coating, nozzle printing, and ink-jet printing method for their advantages of large scaling and relatively inexpensive cost.^{8,9}

A lot of approach ways for WOLED have been reported since the first report of Kido group.¹⁰ Many researchers are also involved in ongoing efforts to make two or three emissive layers in OLED device. Also, because host-dopant system can significantly improve efficiency and show stable color purity, it was often used in WOLED.

Especially, a system that shows high efficiency with a hole or electron transporting material blended with emitting compounds is being actively considered as a cohost system.¹¹ Also, in order to develop the host for

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high performance, many molecules were designed and synthesized.

In this study, T-TATa as new host material with t-butyl group was characterized and compared to TATa without t-butyl group in WOLED. White EML is proposed by using blending and spin-coating method through a co-host/dopant system based on TATa or T-TATa and NPB as co-host.

2. EXPERIMENTAL DETAILS

2.1. General Experiment

TATa, T-TATa and N,N'-Bis(naphthalen-1-yl)-N,N'bis(phenyl)-benzidine [NPB] were used for the host materials used in the emitting layer, and 4,4'-bis[4-(dip-tolylamino)styryl]biphenyl [DPA-VBi] and 5,6,11,12tetraphenylnaphthacene [rubrene] were used as each blue and yellow as the dopant materials. TATa:NPB or T-TATa:NPB was mixed in a mass ratio of 60:40 for host materials and DPAVBi with ratio of 4% and rubrene with ratio of 0.25% were mixed for dopant materials based on the mass of co-host. Solvent was toluene. A water-dispersed PEDOT/PSS mixture (Baytron PVP CH8000, H. C. Starck GmbH) was spin-coated on top of indium tin oxide (ITO) in air for 40 nm thickness. The spin-coated films were baked on a hot plate at 110 °C for 5 minutes in air and 200 °C for 5 minutes

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Figure 1. Chemical structures of host and dopant materials.

in N₂ glove box. The emitting layer was spin-coated to obtain a thickness of 50 nm. The spin-coated films of emitter were baked on a hot plate (110 °C, 30 min) in N₂ glove box. Electron-transporting layers 1,3,5tris(*N*-phenylbenzimidazole-2-yl)benzene (TPBi) with a thickness of 20 nm were deposited by vacuum evaporation (at a pressure about 10⁻⁶ Torr). Also, LiF (1 nm) and Al films were sequentially deposited on the electrontransporting layer. *I*–*V*–*L* characteristics of the WOLED devices and EL spectra were obtained using a Keithley 2400 source measure unit and a Minolta CS-1000A spectrophotometer. The optical absorption spectra were obtained by HP 8453 UV-VIS-NIR spectrometer. Perkin Elmer luminescence spectrometer LS50 (Xenon flash tube) was used for photo spectroscopy (PL).

2.2. Synthesis of TATa and T-TATa

The method of synthesis for TATa in Figure 1 is published in Org. electron.¹² Scheme 1 shows synthetic routes of T-TATa.

In a 250 mL round-bottomed flask, compound (1) (3.4 g, 5.37 mmol), compound (2) (1.62 g, 5.91 mmol), $Pd(OAc)_2$ (36 mg, 0.16 mmol), (o-OMePh)₃P (114 mg, 0.32 mmol), and K₃PO₄ (4.6 g, 21.5 mmol) were placed with anhydrous DME 120 ml and water 30 ml. The temperature was raised to 100 °C. After reaction is completed, it is extracted by chloroform and water. After removing water from the mixture, it was filtered by using MgSO₄. The mixture was columned under chloroform:Hexane (1:3) mixed solvent. The product was concentrated under reduced pressure and re-precipitated with methanol to obtain a yellow solid material (yield: 83.4%).

T-TATa has isomer form. So NMR is not conformed, but EA and mass can be conformed. HRMS: m/z 781.3697 (M+, calcd 781.3709). Anal. Calcd for C60H47N1: C, 92.15; H, 6.06; N, 1.79 Found: C, 92.02; H, 6.04; N, 1.74.

3. RESULTS AND DISCUSSION

Figure 1 shows the chemical structures of host and dopant materials. TATa and T-TATa are blue emitters as host materials. In the case of T-TATa, t-butyl group was included in anthracene core of TATa. NPB is used as a co-host material which has HTL property and wide band gap similar to a blue emitter. DPAVBi and rubrene were each used as blue dopant and yellow dopant.

The optical and electrochemical properties of the synthesized materials were summarized in Table I. Figure 2 exhibits UV-Visible (UV-Vis.) absorption and PL spectra of two compounds in toluene solution and film state. UV-Vis. absorption in solution and film state of two compounds shows the typical anthracene absorption band in range of 350~405 nm. Major absorption peak of TATa and T-TATa in toluene solution state exhibited at 397 nm and 396 nm, respectively. Compared to solution state, UV-Vis. absorption in film state was slightly red shifted as 404 nm and 402 nm, which indicated an increase in conjugation length in the solid state. The increase in conjugation



Scheme 1. Synthetic routes of T-TATa.

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Table 1.	Optical and electrical properties of synthesized materials.						
	Solution ^a		Film ^b				
	UV _{max}	PL _{max}	UV _{max}	PL _{max}	HOMO	LUMO	
	(nm)	(nm)	(nm)	(nm)	(eV)	(eV)	
TATa	382, 397	456	384, 404	466	-5.37	-2.50	
T-TATa	382, 396	456	384, 402	465	-5.28	-2.40	

Table I. Optical and electrical properties of synthesized materials.

Notes: ^aToluene solution $(1 \times 10^{-5} \text{ M})$. ^bFilm on glass.

length might be due to the conformation of solid state π -stacked compounds being more planar.¹³ The PL maximum wavelength of TATa and T-TATa were also measured at 456 nm in solution state, 466 nm and 465 nm in film state, respectively. TATa and T-TATa including t-butyl group were observed similar optical maximum values of UV-Vis. absorption and PL spectra.

Optical band gap obtained from onset of UV absorption was 2.87 eV for TATa and 2.88 eV for T-TATa. The value of highest occupied molecular orbital (HOMO) level was calculated with the use of cyclovoltammetry, and lowest unoccupied molecular orbital (LUMO) level with the use of HOMO level and optical band gap. HOMO-LUMO values of T-TATa have at -5.28 eV and -2.40 eV slightly higher values than -5.37 eV and -2.50 eV of TATa.

 Table II. EL performances of the WOLED devices using synthesized materials at 10 mA/cm².

	L. E. ^{<i>a</i>} (Cd/A)	P. E. ^b (lm/W)	CIE(x, y)	EL _{max} (nm)
ТАТа	1.58	1.28	(0.30, 0.39)	466, 550
T-TATa	3.39	2.68	(0.28, 0.38)	468, 550

Notes: ^aLuminance efficiency. ^bPower efficiency.

It may be due to the donating effect of t-butyl group in T-TATa.

The device structure was fabricated as ITO/PEDOT (40 nm)/emitting layer: TATa or T-TATa + NPB + dopants (50 nm)/TPBi (20 nm)/LiF (1 nm)/Al. Device performances of WOLED using TATa or T-TATa are summarized in Table II. WOLED devices were fabricated by spin-coating method using co-host of TATa or T-TATa and NPB as well as dopants. Figure 3 shows the luminance efficiency (cd/A) versus current density of TATa and T-TATa. WOLED devices based on TATa or T-TATa exhibited luminance efficiencies of 1.58 cd/A and 3.39 cd/A at 10 mA/cm², respectively. Power efficiencies (lm/W) were



Figure 2. UV-Visible absorption and PL spectra of TATa and T-TATa: (a) solution in toluene $(1.0 \times 10^{-5} \text{ M})$, (b) spin-coated film.

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Figure 3. (a) Luminance efficiency and (b) power efficiency of WOLED devices.



Figure 4. EL spectra of WOLED devices.

4. CONCLUSION

1.28 lm/W and 2.68 lm/W. T-TATa in which t-butyl group was added at 2-position of anthracene showed higher efficiency than TATa because t-butyl group can help to prevent $\pi - \pi^*$ stacking interaction between adjacent molecules in spin-coated film.¹⁴ As shown in Figure 4, the EL maximum values of TATa and T-TATa were exhibited at 466 nm or 468 nm of blue emission and 550 nm of orange emission, respectively. In case of the CIE values, similar color coordinates of TATa and T-TATa were shown at (0.30, 0.39) and (0.28, 0.38). Delivered by Publishing Technology

WOLED devices were fabricated by spin-coating method

based on co-host such as TATa or T-TATa with NPB.

Also, DPAVBi and rubrene were used as blue and orange

dopants. As a result, the device used T-TATa including t-

butyl group as the co-host showed luminance efficiency of

3.39 cd/A, which is about two times as high as 1.58 cd/A

of TATa. CIE values of TATa and T-TATa device were

(0.30, 0.39) and (0.28, 0.38), respectively. In conclusion, WOLED device based on solution process including host material, a compound that includes substitution of t-butyl group into the anthracene core group showed higher performance than another compound device without t-butyl group.

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